

**This Page Is Inserted by IFW Operations
and is not a part of the Official Record**

BEST AVAILABLE IMAGES

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- **BLACK BORDERS**
- **TEXT CUT OFF AT TOP, BOTTOM OR SIDES**
- **FADED TEXT**
- **ILLEGIBLE TEXT**
- **SKEWED/SLANTED IMAGES**
- **COLORED PHOTOS**
- **BLACK OR VERY BLACK AND WHITE DARK PHOTOS**
- **GRAY SCALE DOCUMENTS**

IMAGES ARE BEST AVAILABLE COPY.

**As rescanning documents *will not* correct images,
please do not report the images to the
Image Problem Mailbox.**

PATENT SPECIFICATION

DRAWINGS ATTACHED

1037423

1037423



Date of Application and filing Complete Specification May 13, 1963.

No. 18787/63.

Application made in Sweden (No. 5589) on May 18, 1962.

Complete Specification Published July 27, 1966.

© Crown Copyright 1966.

Index at acceptance:—H1 BF

Int. Cl.:—H 01 m 27/60

COMPLETE SPECIFICATION

Method of Supplying a Fuel Cell with Fuel in the form of Hydrogen

We, ÄLLMÄNNA SVENSKA ELEKTRISKA AKTIEBOLAGET, a Swedish Company of Västerås, Sweden, do hereby declare the invention, for which we pray that a Patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following Statement:—

Fuel cells employing hydrogen as the combustible substance and oxygen or air as the oxidising substance have been the object of great interest, not least for use on ships because of the good working times given by fuel cell apparatus constructed from them. The need to keep large amounts of hydrogen is however a very serious disadvantage with the use of such fuel cells, especially because of the danger of explosion this involves and the large storage space needed. This disadvantage is present not only when the fuel cells are used on ships but also when they are used in confined spaces as reserve power aggregates.

The use of ammonia has been proposed as the active combustible substance in fuel cells. The use of ammonia as a fuel should have great advantages, especially in cases where limited space is available for storage of the fuel. As ammonia may be kept simply in liquid form, it requires only small storage vessels so that storage space and freight costs will be small. Ammonia is moreover a cheap fuel. In spite of these favourable properties, however, ammonia has not been used in practice as a fuel in fuel cells. This is connected with difficulties in producing a sufficiently active electrode material, which can effectively cause the necessary oxidation of the ammonia.

The present invention relates to a method of supplying fuel to a fuel cell which avoids the disadvantages connected with storage of

large amounts of hydrogen but which takes advantage of the use of ammonia without the attendant disadvantages associated with the earlier proposed use of ammonia.

According to the invention a method of supplying a fuel cell with fuel in the form of hydrogen, which is supplied to a hydrogen chamber arranged in the fuel cell, is characterised in that the hydrogen chamber of the fuel cell is connected to an ammonia cracker comprising a reaction chamber for cracking the ammonia and a combustion chamber for heating the reaction chamber, from which hydrogen and nitrogen formed in the reaction chamber are led to the hydrogen chamber of the fuel cell where the hydrogen is partly consumed for the fuel cell operation, and that hydrogen not consumed in the fuel cell together with the nitrogen are led to the combustion chamber of the ammonia cracker and are there used as fuel.

By the invention use is made of the advantage of ammonia from a storage point of view and also of the large reactivity of hydrogen which is superior to that of ammonia. The disposal of the hydrogen in the method according to the invention is of particularly great importance. A necessary condition for the efficient working of a fuel cell employing a fuel consisting of hydrogen and nitrogen is that the percentage of hydrogen is high. With a low percentage of hydrogen the efficiency is poor and the cell voltage low. Since the gas mixture coming from the cracker is supplied to the hydrogen chamber of the fuel cells, and the gas led away from the hydrogen chamber, in which gas the hydrogen percentage is substantially lower, is not used as fuel in the fuel cell but as a source of energy for the ammonia cracking, maximum use can be made of the hydrogen. Upon combustion of the hydrogen in the

combustion chamber of the cracker the hydrogen is thus used completely even if its percentage in the gas mixture is low. In order to avoid using an extra heating means in the cracker, it is especially suitable to adapt the supply of the hydrogen and nitrogen to the fuel cell hydrogen chamber and their discharge therefrom so that the amount of hydrogen not consumed in the fuel cell, which is used as fuel in the combustion chamber of the ammonia cracker, gives just that energy which is necessary so that the cracking shall continue during maintenance of the desired gas supply to the hydrogen chamber of the fuel cell. In the majority of practical cases this means that somewhat more than two-thirds of the amount of hydrogen supplied to the fuel cell is used there and somewhat less than one third of it is used as fuel in the combustion chamber of the ammonia cracker.

The method according to the invention may be used with advantage for the operation of fuel cell apparatus, e.g. for submarines, with simultaneous supply of the fuel cell apparatus with oxygen from a container containing liquid oxygen and possibly liquid nitrogen. Fuel cells are then used which comprise an oxygen chamber, to which oxygen is led which suitably is only partly consumed for operation of the fuel cell. According to one embodiment of the invention the hydrogen led to the combustion chamber of the ammonia cracker is burnt with unconsumed oxygen from the oxygen chamber of the fuel cell, and at least a part of the exhaust from the combustion chamber of the ammonia cracker is led after removal of occurring water vapour to the container with liquid oxygen and possibly liquid nitrogen, where it mixes with the contents of the container with the giving up at least a part of the nitrogen of the exhaust and the taking up oxygen present in the container, after which the gas enriched with oxygen is led to the oxygen chamber of the fuel cell and is there used as oxidising agent. Nitrogen formed in the ammonia cracker or at least the main part of it is thereby collected in the container. It is particularly advantageous to adapt the exhaust supply to the container so that the amount of nitrogen given up by the exhaust in the container corresponds to the amount of new nitrogen produced simultaneously in the ammonia cracker.

According to another embodiment of the invention the hydrogen led to the combustion chamber of the ammonia cracker is burnt with unconsumed oxygen from the oxygen chamber of the fuel cell and at least a part of the exhaust from the combustion chamber of the ammonia cracker is led after removal of occurring water vapour to a condenser, which is cooled with liquid oxygen of lower pressure than the pressure in the

exhaust, at least a part of the nitrogen in the exhaust then being condensed with simultaneous vaporising of the oxygen used as cooling medium which is led to the oxygen chamber of the fuel cell, possibly mixed with non-condensed nitrogen, and is there used for supplying the fuel cell with oxygen. It is particularly advantageous to adapt the amount of nitrogen condensed in the condenser so that it corresponds to the amount of new nitrogen produced simultaneously in the ammonia cracker. A special advantage of the mentioned embodiment of the invention is that they can be used for renewal of the atmosphere in the submarine.

Oxygen maintenance to the oxygen chamber of the fuel cell in submarines may also occur by means of oxygen recovery from sea water. The hydrogen led to the combustion chamber of the ammonia cracker is thereby burned up as in the above mentioned cases with unconsumed oxygen from the oxygen chamber of the fuel cell and the exhaust from the combustion chamber of the ammonia cracker is led to a gas exchanger, where it comes into contact with sea water passing through the gas exchanger with the giving up a part of the nitrogen of the exhaust and the taking up oxygen dissolved in the sea water, after which the gas enriched with oxygen is led to the oxygen chamber of the fuel cell and is there used as an oxidising agent. The exhaust from the combustion chamber of the cracker consists mainly of nitrogen gas. The condition for oxygen being taken up at the contact with the sea water is that only a part of the nitrogen is dissolved so that there is always nitrogen present in the gas exchanger, with which the oxygen may be mixed and conveyed out of the gas exchanger. For oxygen recovery nitrogen is thus used which has less solubility in water than oxygen. The principle of using oxygen dissolved in sea water as oxidising agent is known for combustion motors in submarines. The combustion gases were used for oxygen recovery and it was presumed that a condition for the functional efficiency of the method should be that the combustion gases should have a greater solubility in water than oxygen.

The invention will now be described in greater detail, by way of example, with reference to the accompanying drawing, in which

Figure 1 illustrates schematically an arrangement for carrying out the method according to the invention,

Figure 2 shows schematically an arrangement for using the method in a submarine with an oxygen supply from a container containing liquid oxygen, with which nitrogen is brought into direct contact,

Figure 3 shows schematically a modification of the arrangement according to Figure 2, in which the oxygen maintenance is arranged

by evaporation of oxygen during condensation of the nitrogen of the exhaust in a condenser where the media are in indirect contact with each other, and

5 Figure 4 illustrates schematically an arrangement for using the method in a submarine with recovery of oxygen from the sea water, with which the exhaust is brought into direct contact.

10 The arrangement according to Figure 1 comprises in principle three units, namely a tank 10 containing liquid ammonia, an ammonia cracker 11 and a fuel cell 12. The ammonia cracker comprises a reaction chamber, e.g. consisting of several tubes or retorts 15 13a containing a suitable cracking catalyst with for example iron oxide, nickel or platinum as active component and a combustion chamber 14 surrounding the tubes 13a. The fuel cell contains two porous electrodes 15 and 16 between which is the electrolyte 17 in an electrolyte chamber 18. Outside the electrode 15 is a gas chamber 19 for the fuel and outside the electrode 16 a gas chamber 20 for the oxidising medium. The electrode 15 may for example consist of an active nickel electrode produced in a known way by pressing and sintering a powder mixture of aluminium nickel alloy and carbonyl nickel, after which the sintered product is activated by removing of the aluminium with alkali. The electrode 16 may for example consist of a silver electrode produced in an analogous and likewise known way 30 from a powder mixture of silver-aluminium alloy and carbonyl nickel. The electrolyte 17 may consist, for example, of potassium hydroxide solution. The operating temperature may be about 80°C. When the arrangement is in operation the ammonia is conveyed from the tank 10 via a conduit 21 to the cracker 11 where two volumes of ammonia are cracked with the formation of one volume of nitrogen and three volumes of hydrogen. 45 The mixture of nitrogen gas and hydrogen gas is fed via a conduit 22, which may contain a small buffer tank 23 and a control valve 24, to the gas chamber 19 of the fuel cell where a major part, preferably about two-thirds of the hydrogen, is consumed by oxidation during the electrode reaction taking place in the fuel cell. The remaining hydrogen gas together with the unchanged nitrogen gas passes via a conduit 25, which may contain a small buffer tank 26 and a control valve 27, to the combustion chamber 14 of the cracker 11, where it is burnt up by air or oxygen gas, which may be supplied via a conduit 28, before the gases reach the cracker. Alternatively, the combustion chamber may lie outside the actual cracker, the warm combustion gases being led from the combustion chamber into the actual cracker in order to heat the gas in the reaction chamber. The combustion gases,

which consist of water vapour, nitrogen gas and possibly excess oxygen gas are led away via a conduit 29. If the arrangement is used in a submarine, the nitrogen gas may with advantage be condensed so that the presence of the submarine is not revealed by gas bubbles. If such a large hydrogen consumption is allowed in the fuel cell that the amount of unconsumed hydrogen gas is insufficient to maintain the necessary cracking of the ammonia, the cracker may be equipped with an extra heating arrangement, e.g. consisting of electric resistance coils. The oxidising medium may consist of oxygen gas or air which is led to the oxygen chamber 20 via a conduit 30. Nitrogen gas which may be present and possible excess oxygen gas are led away via a conduit 31.

In the arrangement according to Figure 2, which partly comprises the same units as the arrangement of Figure 1, the exhaust from the combustion chamber 14 of the cracker is led via the conduit 29 to a condenser 32 for removal of the water vapour content in the exhaust. The exhaust passes a drying device 36, e.g. comprising molecular filters or silica gel for removal of remaining traces of water vapour and other absorbable material, before being transported via a conduit 33 with a valve 34 to a container 35 containing liquid oxygen and possibly liquid nitrogen. The exhaust may be led directly into the container 35, e.g. via one or more perforated inlet pipes 37 arranged at the bottom of the container. In the container 35 at least a part of the nitrogen gas is dissolved, oxygen gas being simultaneously vapourised. After being heated in a heat exchanger 39, which is used simultaneously for cooling the nitrogen gas led to the container 35, the oxygen-enriched gas passes via conduits 38 and 30 to the oxygen chamber 20 of the fuel cell, where the oxygen gas is partly consumed. Unconsumed oxygen gas is led via the conduit 31 to the combustion chamber 14 of the ammonia cracker where it acts as oxidising medium for the hydrogen gas from the hydrogen chamber 19 of the fuel cell. In order to safely maintain the nitrogen gas balance in the system, so that the amount of nitrogen gas which remains in the container 35 is as large as that produced simultaneously in the cracker 11, a part of the exhaust gas from the container 35 may be condensed in a liquefier 40. The container 35 contains a heating coil 41 for the leading there-through of a heating medium so that the necessary temperature and thus pressure may be maintained in the tank. Adjustment of the amount of oxygen gas in the conduit 30 occurs through evaporation of oxygen from a tank 42 with a heating coil 43. The tank 42 contains pure oxygen and the oxygen is led away via a conduit 44 to the conduit 30.

For renewal of the atmosphere in the submarine used air may be taken in via conduit 45. Simultaneously, oxygen rich gas from a conduit 46 with a valve 47 is mixed with oxygen-poor gas from a conduit 48 with a valve 49 and the mixture is led out into the submarine via a conduit 50 so that pressure and composition of the atmosphere in the submarine and kept constant. The impurities in the used air are burnt in the ammonia cracker.

Figure 3 shows a modification of part of the arrangement of Figure 2, items 35, 37, 38, 40, 41, 42, 43 and 44 of Figure 2 being replaced while the remaining items are the same. According to Figure 3 the exhaust coming through the conduit 33 is led through a condenser 51 where at least a part of the nitrogen gas is condensed and collected in a container 52. Uncondensed exhaust is led via a conduit 53 to the conduit 30. The condensation of the nitrogen gas is effected by liquid oxygen, which is kept at a lower pressure than the exhaust so that its boiling point is below the boiling point for the nitrogen, being led through a cooling coil 54 in the condenser. The lower pressure in the cooling coil is maintained by means of a pump 55 and is regulated by a valve 56. Vaporised oxygen gas is separated from liquid oxygen in a separating vessel 57. The liquid oxygen is supplied to the cooling coil 54 from an oxygen tank 58 provided with a heating coil 59 via a conduit 60 containing a valve 61. The valve 61 is controlled by the liquid level in the separating vessel 57, so that complete evaporation of the liquid in the separating vessel 57 is prevented. The conduit 30 may also be supplied with oxygen gas direct from the tank 58 via a conduit 62 containing a valve 63. In order to maintain the nitrogen gas balance in the system, so that the amount of nitrogen gas which remains in the container 52 is as large as the amount of new nitrogen produced simultaneously in the cracker 11, the temperature of the oxygen in the cooling coil 54 and thereby the cooling effect is regulated by adjusting the pressure in the separating vessel 57 with the valve 56. Adjustment of the amount of oxygen gas in the conduit 30 occurs by means of evaporation of oxygen from the tank 58 with the heating coil 59. At very low power output from the fuel cell battery heat leakage into the system may be so high that the amount of oxygen gas which escapes in the conduit 30 exceeds that consumed by the fuel cell and the ammonia cracker, with the result that the oxygen pressure would increase in the system. This may be prevented by using a liquefier 64.

In the arrangement according to Figure 4, which partly comprises the same units as the arrangement of Figure 1, the exhaust

from the combustion chamber 14 of the cracker is led via the conduit 29 to a gas exchanger 72. This may for example consist of an open longitudinal channel arranged in or near the bottom of the submarine, which takes up a considerable part of the cross-section of the boat. Since large amounts of water and gas must be mixed because of the low oxygen content of the water, the volume of the channel is thus considerable. When the boat is in motion, sea water enters the gas exchanger at its forward end 73a and leaves it after passage through it at its stern end 74a. The gas from the conduit 29 enters for example through nozzles in the gas exchanger at the forward end 73b of the boat. In the gas exchanger a part of the nitrogen of the exhaust is dissolved in the water, while oxygen gas dissolved in the water is taken up by nitrogen gas pockets during their transport aft, which then consequently do not completely dissolve. The gas enriched with oxygen gas is taken out of the exchanger and into the boat via a valve arrangement in the stern end 74b of the boat, which arrangement lets in the gas but keeps out the water. The oxygen-rich gas then passes via the conduit 30 to the oxygen chamber 20 of the fuel cell, where the oxygen gas is partly consumed. Unconsumed oxygen gas is led via the conduit 31 to the combustion chamber 14 of the ammonia cracker, where it acts as an oxidising agent for the hydrogen gas from the hydrogen gas chamber 19 of the fuel cell. If the gas exchanger is dimensioned so that the amount of nitrogen gas given up by the exhaust therein is as large as the amount of new nitrogen gas produced simultaneously in the ammonia cracker, the nitrogen gas balance in the system can be maintained. The gas exchange cannot in general be carried out so that the requirement for oxygen gas is exactly covered by the amount taken up from the water. In general the taking up of oxygen gas is the more effective process, and material balance is then easily effected by means of recirculation of the gas from the exchanger via a conduit 75 with a control valve 76. With the especially advantageous conditions referred to in the description of Figure 1, i.e. that the amount of hydrogen not consumed in the fuel cell, which is used as fuel in the combustion chamber of the ammonia cracker, given just the energy necessary so that the cracking shall continue during maintenance of the desired gas supply to the hydrogen chamber of the fuel cell, the conditions will be as follows with the case described in Figure 4 as example:—Cracking of 2 volumes ammonia gas in the cracker 11 gives 1 volume of nitrogen gas and 3 volumes of hydrogen gas in the conduit 22. Of this about 2 volumes of hydrogen gas is consumed in the fuel cell and therefore

about 1 volume of nitrogen gas and about 1 volume of hydrogen gas passes to the conduit 25. From the conduit 31 comes nitrogen gas, e.g. 1 volume, and about $\frac{1}{2}$ volume of oxygen gas which is required for the combustion of the hydrogen gas in the cracker. In the conduit 29 the combustion gas then consists of about 2 volumes nitrogen gas and about 1 volume of water vapour, with a little excess oxygen. In the gas exchanger 72 the gas gives up the vapour and further nitrogen gas and takes up oxygen gas. The gas exchanger 72 is so dimensioned, and the adjusting conduit 75 connected to it is so adjusted, that the total result of the exchanging in the gas exchanger and the adjusting conduit is that about 1 volume nitrogen is given up and about $1\frac{1}{2}$ volumes oxygen gas are taken up by the gas. In the conduit 30 the gas then consists of about 1 volume of nitrogen gas and about $1\frac{1}{2}$ volumes of oxygen gas, which is necessary if the oxygen gas shall be sufficient to consume the stated amount of about 2 volumes of hydrogen gas in the fuel cell and oxidise the stated amount of about 1 volume of hydrogen gas in the combustion chamber of the cracker. The amount of nitrogen gas passing through the oxygen chamber 20 of the fuel cell and through the conduits 25 and 30 is not critical, but should not be too large in order to avoid blocking of the electrode pores with the inert nitrogen gas. Analogous conditions apply for the embodiments according to Figures 2 and 3. Consequently it follows that the oxygen gas supply through the conduit 30 is adapted so that the oxygen is at least sufficient for the reaction with the hydrogen in the fuel cell and in the combustion chamber of the ammonia cracker.

WHAT WE CLAIM IS:—

1. Method of supplying a fuel cell with fuel in the form of hydrogen, which is supplied to a hydrogen chamber arranged in the fuel cell, characterised in that the hydrogen chamber of the fuel cell is connected to an ammonia cracker comprising a reaction chamber for cracking the ammonia and a combustion chamber for heating the reaction chamber, from which hydrogen and nitrogen formed in the reaction chamber are led to the hydrogen chamber of the fuel cell where the hydrogen is partly consumed for the fuel cell operation, and that hydrogen not consumed in the fuel cell together with the nitrogen are led to the combustion chamber of the ammonia cracker and there used as fuel.

2. Method according to claim 1 in which supply of the hydrogen and nitrogen to the hydrogen chamber of the fuel cell and their discharge therefrom are adjusted so that the amount of hydrogen not consumed in the fuel cell, which is used as fuel in the com-

bustion chamber of the ammonia cracker during combustion, gives just the energy necessary for the cracking therein.

3. Method according to claim 1 or 2 for supplying a fuel cell in for example a submarine, which fuel cell comprises an oxygen chamber arranged in the fuel cell and supplied with oxygen which is partly consumed for the fuel cell operation, in which the hydrogen led to the combustion chamber of the ammonia cracker is burned with unconsumed oxygen from said oxygen chamber, and in which at least a part of the exhaust from the combustion chamber of the ammonia cracker after removal of occurring water vapour is led to a container containing liquid oxygen and possibly liquid nitrogen, where the exhaust is mixed with the contents of the container with the giving up of at least a part of the nitrogen of the exhaust and the taking up of oxygen present in the container and the gas enriched with oxygen is led to said oxygen chamber, whereby the oxygen of the container is used for supplying the fuel cell with oxygen and nitrogen formed in the ammonia cracker or at least the main part of it is collected in the container.

4. Method according to claim 3, in which the amount of nitrogen given up in the container corresponds to the amount of new nitrogen produced simultaneously in the ammonia cracker.

5. Method according to claim 1 or 2 for supplying a fuel cell in for example a submarine, which fuel cell comprises an oxygen chamber arranged in the fuel cell and supplied with oxygen which is partly consumed for the fuel cell operation, in which the hydrogen led to the combustion chamber of the ammonia cracker is burned with unconsumed oxygen from said oxygen chamber, and in which at least a part of the exhaust from the combustion chamber of the ammonia cracker after removal of occurring water vapour is led to a condenser, which is cooled with liquid oxygen of lower pressure than the pressure in the exhaust, at least a part of the nitrogen in the exhaust then being condensed during simultaneous vaporising of the oxygen used as cooling medium, which is led to said oxygen chamber, possibly mixed with non-condensed nitrogen, and is there used for supplying the fuel cell with oxygen.

6. Method according to claim 5, in which the amount of nitrogen condensed in the condenser corresponds to the amount of new nitrogen produced simultaneously in the ammonia cracker.

7. Method according to claim 1 or 2 for supplying a fuel cell in a submarine, which fuel cell comprises an oxygen chamber arranged in the fuel cell and supplied with oxygen which is partly consumed for the fuel cell operation, in which the hydrogen

- led to the combustion chamber of the ammonia cracker is consumed with unconsumed oxygen from said oxygen chamber, and in which the exhaust from the combustion chamber of the ammonia cracker is led to a gas exchanger, where it is brought into contact with sea water passing through the gas exchanger with the giving up of a part of the nitrogen of the exhaust and the taking up of oxygen dissolved in the sea water, and the gas enriched with oxygen is led to said oxygen chamber, whereby oxygen gas dissolved in the sea water is used for supplying the fuel cell with oxygen.
- 5 3. Method according to claim 7, in which the gas exchanger is dimensioned so that amount of nitrogen given up therein at least corresponds to the amount of new nitrogen produced simultaneously in the ammonia cracker.
- 10 4. Method according to claim 7 or 8, in which the amount of oxygen in the gas enriched with oxygen led to said oxygen chamber is adjusted so that the volume of the oxygen taken up in the gas exchanger and transported to the oxygen chamber is one and a half times as great as the volume of nitrogen simultaneously dissolved by the sea water in the gas exchanger.
- 15 5. Method according to any one of claims 3 to 9, in which air from the living-rooms of the submarine together with unconsumed oxygen from said oxygen chamber is led through the combustion chamber of the ammonia cracker for combustion of vitiating compounds in the air.
- 20 6. A method of supplying a fuel cell with fuel constructed and arranged substantially as herein described with reference to any one of Figures 1 to 4 of the accompanying drawings.
- 25 7. J. Y. & G. W. JOHNSON,
Furnival House, 14—18, High Holborn,
London, W.C.1.
Chartered Patent Agents,
Agents for the Applicants.
- 30 8. Method according to any one of claims 3 to 9, in which air from the living-rooms of the submarine together with unconsumed oxygen from said oxygen chamber is led through the combustion chamber of the ammonia cracker for combustion of vitiating compounds in the air.
- 35 9. A method of supplying a fuel cell with fuel constructed and arranged substantially as herein described with reference to any one of Figures 1 to 4 of the accompanying drawings.
- 40

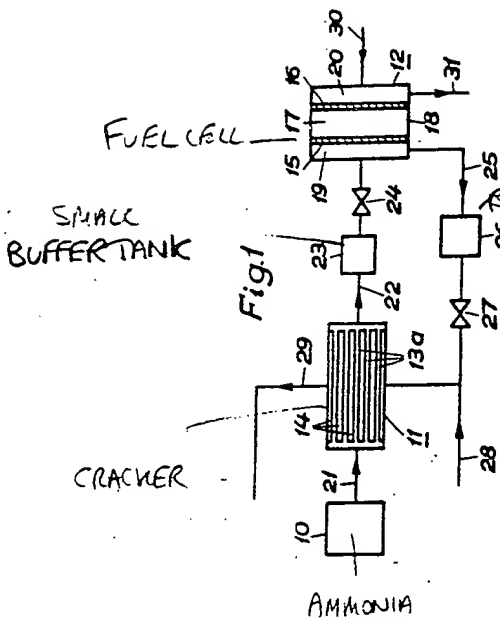


Fig. 4

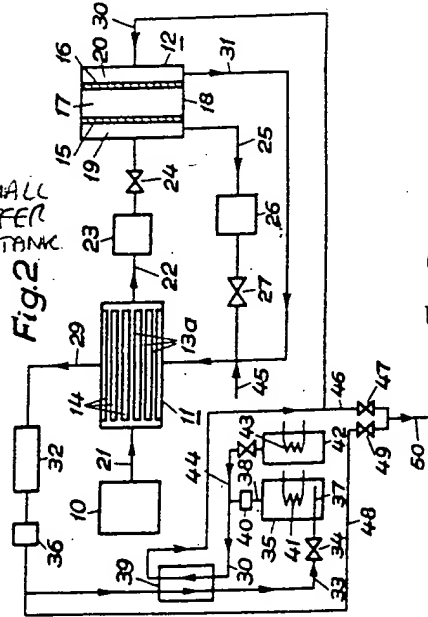
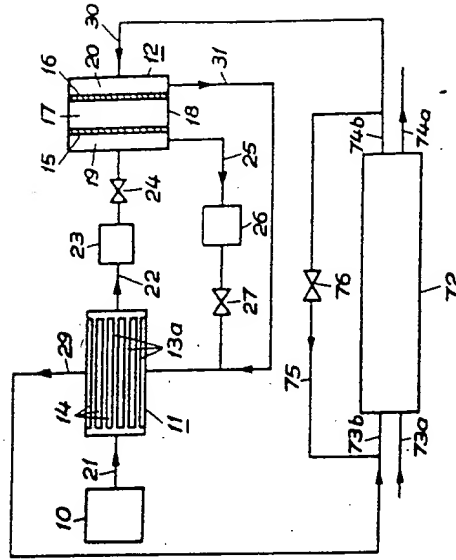


Fig. 3

